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## (54) Tobacco smoke filter

(57) Volatile oxides are removed from tobacco smoke by means of a metal co-ordination complex which is relatively stable in air but which is active in contact with tobacco smoke to result in removal of CO and/or NO from the smoke. Such complexes are suitably oximic complexes of metals having two or more valence states, e.g. ferrous and ferric complexes with  $\alpha$ -naphthaquinone-2-oxime and  $\beta$ -naphthaquinone-1-oxime. The complex or complexes, suitably supported by a particulate carrier, can be incorporated in tobacco smoke filters, e.g. cigarette filters.

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The claims were filed later than the filing date within the period prescribed by Rule 25(1) of the Patents Rules 1982.  
The matter shown in the printed specification between square brackets was furnished after the filing date of the application, the application as filed being defective as regards this matter.

SPECIFICATION  
Tobacco Smoke Filter

The present invention relates to the chemical removal of gaseous components from tobacco smoke, and to filters for this purpose.

5 The removal of gaseous tobacco smoke components such as carbon monoxide (CO) and nitric oxide (NO) has been the subject of much attention for a considerable time; removal of such components by chemical reaction has seemed an attractive solution, but despite the expenditure of much effort in this direction, there has been no real practical or commercial success in developing chemical means for this purpose. 5

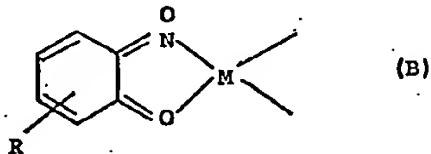
10 Prior chemicals suggested as tobacco smoke filter additives for removal of gases such as CO from the smoke have one or more of the following major disadvantages; they are very reactive and so are readily oxidised, thereby rapidly losing their reactivity on exposure to air; they have a high molecular weight to reactive site ratio, i.e. there is a large redundant mass associated with each reaction site; they may become inactive through interaction with other components of tobacco smoke; they may be indirectly or directly 10

15 poisonous to the smoker. 15

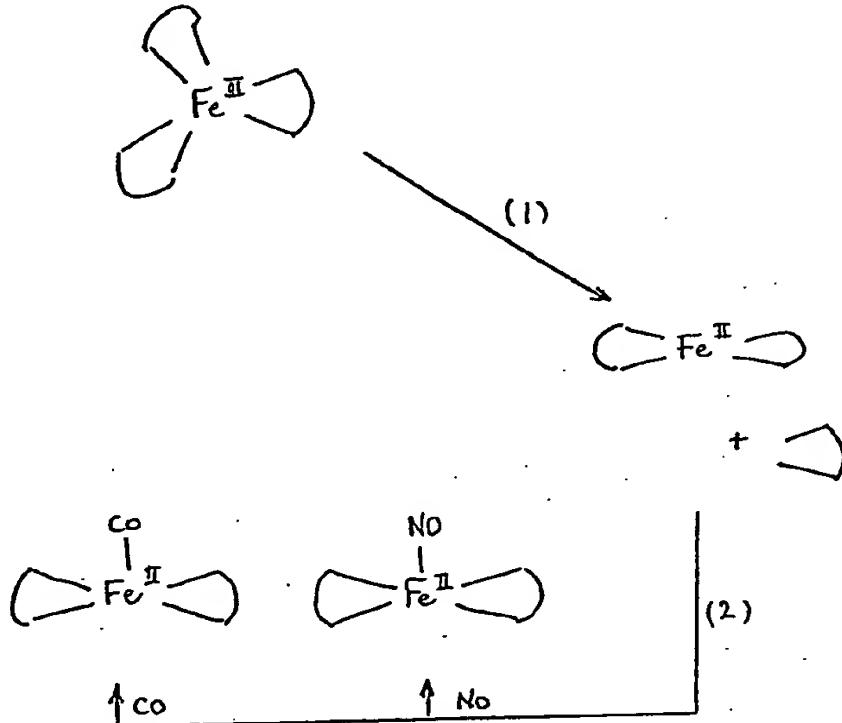
The present invention resides in the removal of CO and/or NO from tobacco smoke by reaction with a metal co-ordination complex (e.g. an oximic complex and preferably an oximic complex of the type B below) which is relatively stable in air but which is active in contact with tobacco smoke to result in removal of CO and/or NO from the smoke. The metals of the complexes used according to the invention are 20

20 generally transition or redox or like metals exhibiting more than one valency. 20

The above-mentioned oximic complexes B may be generally represented by the formula:



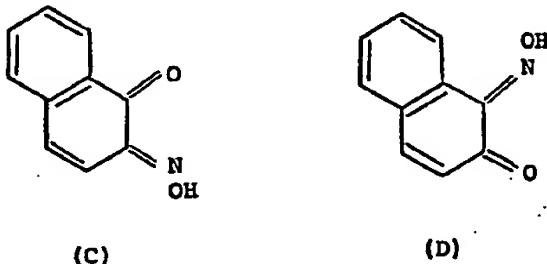
wherein R is an aliphatic or organic radical of relatively low molecular weight (e.g. phenyl, benzyl, etc) and M is a metal (e.g. iron, copper etc) having two or more valence states. These compounds are particularly 25 suitable since the stable complex starting material may be reduced by Lewis base components of the smoke (e.g. nicotine) to produce a reactive species in situ, ready to bind the volatile oxide molecules. This smoke-activation process is shown schematically below using an iron III complex as the example:



wherein;

- step (1) is a fast stoichiometric reaction with Lewis base components of tobacco smoke to form a reactive intermediate and a ligand radical in situ; and
- step (2) is a fast stoichiometric reaction with CO or NO to form a stable complex.

5 Two particular examples of compounds for use according to the invention are the iron complexes 5 derived from  $\alpha$ -naphthaquinone-2-oxime (C) and  $\beta$ -naphthaquinone-1-oxime (D)



10 Complexes of both Fe<sup>II</sup> and Fe<sup>III</sup> co-ordinated with oxime ligands have the following advantageous features: i) they react with CO and/or NO under smoking conditions at a rate sufficient to cause significant reduction in volatile oxide concentration in tobacco smoke; (ii) they are essentially stable towards oxidation 10 and do not decompose significantly when stored in the solid state; (iii) their effectiveness is enhanced rather than reduced by contact with major components of tobacco smoke; (iv) the oxime complexes and the reaction products after interaction with tobacco smoke are involatile. Like complexes with other transition metals, copper etc., can have similar advantages.

15 The invention also provides a tobacco smoke filter, e.g. a cigarette filter, including a metal co-ordination 15 complex as described above for the chemical removal of volatile oxides e.g. CO and/or NO from the tobacco smoke.

20 In practice it is convenient to incorporate the additive compound in the filter upon an essentially inert substrate, preferably one having a high surface area to density ratio. For example, the complexes 20 previously mentioned may be deposited (suitably from solution) on substrate granules of e.g. pumice, silica gel, magnesium silicate etc. The supported or unsupported complex may be disposed in a filter cavity and/or deposited on or distributed through conventional filter material.

25 The invention also provides, for incorporation in tobacco smoke filters, an additive comprising a metal co-ordination complex as described above deposited on a particulate carrier.

25 The invention is illustrated by the following Examples (which are according to the invention) and Controls (which are not according to the invention). 25

#### EXAMPLES 1—4

In these Examples the additive compound used was an iron complex of compound C described above and is referred to as Fe(C)<sub>3</sub>. The complex was deposited onto particulate pumice from solution in acetone. 30

30 About 0.1 g of the pumice bearing about 16 mg of complex was used in each filter. Each filter was a wrapped composite rod 25 mm in length and 8 mm diameter comprising a bonded cellulose acetate tow filter plug spaced apart from a similar plug with the supported complex disposed in the cavity between these elements. Control filters contained pumice treated with acetone containing no complex and dried.

35 Filters were attached to standard tobacco rods and the resulting filtered cigarettes were smoked using a standard smoking machine and the total CO delivered per tobacco rod was measured. The results are 35 shown in Table 1. It is clear from the Table that the complex Fe(C)<sub>3</sub> supported on pumice is effective in removing CO from tobacco smoke.

40 In Table 1 each Example (and each corresponding Control) involved five runs 1—5, and each such run involved the simultaneous smoking in parallel of five of the filter cigarettes with the total smoke delivery being collected and the total CO measured and divided by five to give the CO delivery per rod quoted for each Run. 40

TABLE I  
Effect of Fe complex of compound C on carbon monoxide obtained from tobacco smoke

RUN	CO Delivery (mg) per tobacco rod					Wt. Difference Between Control & Example mg CO	% Difference Between Control and Example
	1	2	3	4	5		
Example 1 Control	14.4 16.1	14.84 16.28	15.00 16.94	14.99 16.44	15.08 16.24	14.86 16.40	1.54 9.4
Example 2 Control	15.24 16.22	14.84 16.06	14.96 16.76	15.36 16.94	15.06 16.26	15.09 16.45	1.36 8.24
Example 3 Control	16.04 17.44	15.52 17.56	16.22 18.06	15.98 17.58	16.12 17.86	15.98 17.7	1.72 9.7
Example 4 Control	15.48 16.8	15.3 17.24	15.48 16.82	15.48 16.79	15.5 16.8	15.46 16.89	1.43 8.4

## EXAMPLES 5—6

In these Examples "MYRIA" (Registered Trade Mark) paper filters made from longitudinally corrugated gathered paper were each impregnated with about 16 mg of an iron complex of compound C or D, referred to as  $\text{Fe}(\text{C})_3$  and  $\text{Fe}(\text{D})_3$ , from solution in chloroform, dried, and tested in a similar way to that used for

5 Examples 1—4. The control filters were "MYRIA" filters treated with chloroform alone and then dried. The 5 results of these tests are shown in Table 2.

## EXAMPLES 7—8

In these Examples iron complexes of compounds C and D, referred to as  $\text{Fe}(\text{C})_3$  and  $\text{Fe}(\text{D})_3$ , were deposited on particulate silica gel from solution as in Examples 1—4; similar control filters used silica gel

10 treated with solvent alone and dried. The filters were tested as for Examples 1—4 and the results are shown 10 in Table 2.

As in Table 1 each CO delivery per rod quoted in Table 2 is obtained by smoking five filter cigarettes simultaneously and dividing the total CO delivery by five.

TABLE 2

15 Effect of Fe Complexes of Compounds C and D on Carbon Monoxide Obtained from Tobacco Smoke 15

Example	Filter Type	CO Delivery (mg) per Tobacco Rod.	Wt. Difference Between Control and Example (mg CO)	% Difference Between Control and Example
5	Myria/ $\text{Fe}(\text{C})_3$	14.9	1.2	7.4
6	Myria/ $\text{Fe}(\text{D})_3$	15.0	1.1	6.8
	Control	16.1	—	—
7	$\text{Fe}(\text{C})_3/\text{Si}^{\text{O}2}$	14.9	1.5	9.1
8	$\text{Fe}(\text{D})_3/\text{Si}^{\text{O}2}$	14.8	1.6	9.1
	Control	16.4	—	—

## CLAIMS

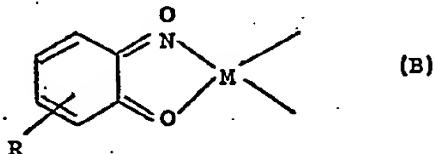
1. A tobacco smoke filter including at least one metal co-ordination complex which is relatively stable in air but which is active in contact with tobacco smoke to result in removal of CO and/or NO from the

20 smoke. 20

2. A tobacco smoke filter according to claim 1 wherein the or each metal is one which exhibits more than one valency.

3. A tobacco smoke filter according to claim 1 or 2 containing a said complex which is an oxime complex.

25 4. A tobacco smoke filter according to claim 1 containing at least one said complex selected from those 25 of formula (B)



wherein R is an aliphatic or aromatic radical and M is a metal having two or more valence states.

5. A tobacco smoke filter according to claim 4 containing at least one said complex selected from 30 ferrous and ferric complexes with  $\alpha$ -naphthaquinone-2-oxime and  $\beta$ -naphthaquinone-1-oxime.

6. A tobacco smoke filter according to any preceding claim wherein the complex is present supported upon a particulate substrate.

7. A tobacco smoke filter substantially as hereinbefore described in any one of the Examples.

8. A method of removing CO and/or NO from tobacco smoke which comprises passing the smoke over 35 a metal co-ordination complex which is relatively stable in air but which is active in contact with tobacco smoke to result in removal of CO and/or NO from the smoke. 35

9. A method according to claim 8 and substantially as hereinbefore described.

10. A metal co-ordination complex supported on a particulate carrier, the complex being one which is relatively stable in air but which is active in contact with tobacco smoke to result in removal of CO and/or NO from the smoke.

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